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TRIPLY DIFFERENTIAL STUDIES OF ATOMIC AND MOLECULAR  
PHOTOIONIZATION USING.. (U) NATIONAL BUREAU OF STANDARDS  
WASHINGTON DC NATIONAL MEASUREME.. J L DEHMER ET AL.

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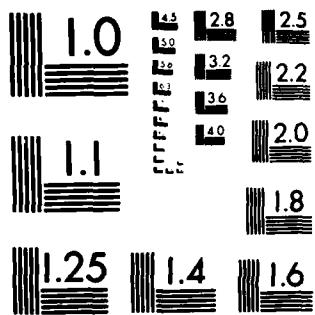
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Basic studies of the dynamics and spectroscopy of atomic and molecular photoionization have been carried out using three experimental probes. The first and most extensively used experimental approach involves triply differential (differential in incident wavelength, electron energy, and ejection angle) photoelectron measurements using synchrotron radiation. Measurements were conducted in the vacuum ultraviolet wavelength range up to $\text{h}\nu \approx 35$ eV on a large variety of atomic and molecular systems. Photoelectron branching ratios		

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20. (Contd.) and angular distributions were obtained for all accessible states. A major emphasis of this work involved the initial exploration of novel effects of autoionization and shape resonances on alternative vibrational ionization channels. The second experimental approach entails measuring the polarization of fluorescence following production of excited molecular ions by photoionization. This experiment allowed the direct measurement of the alignment of molecular ions produced by photoionization and, simultaneously, the branching ratios for degenerate photoelectron channels. New spectroscopic information can also be obtained in this way. The third experimental approach involves measuring photoelectrons in coincidence with photoions. Work to determine the electronic structure of atomic clusters is complete and will be followed by work to measure fragmentation of molecular photoions.

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ANNUAL SUMMARY REPORT

Triply Differential Studies of Atomic and Molecular Photoionization  
Using Synchrotron Radiation (Contract No. N00014-83-F-0005)

Submitted to

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Physics Program Office  
Department of the Navy  
Arlington, VA 22217

Attn: Dr. Bobby R. Junker

Submitted by  
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2. Contract Description

The research covered by this contract involves basic studies of photoionization processes in atoms and molecules using three advanced experimental approaches outlined in Section 4. Using these techniques, new information and insight into the spectroscopy and dynamics of photoionization are obtained by examining several complementary observables, e.g., partial photoionization cross sections, branching ratios, photoelectron angular distributions, alignment of molecular photoions, and fragmentation of molecular ions.

3. Scientific Problem

This study is aimed at resolving roughly four unknown or incompletely known aspects of this problem area: First, this program seeks to characterize major aspects of photoionization dynamics, such as the effects of shape resonances and autoionizing resonances on alternative ionization channels, which can only be studied in a definitive way with the advanced, triply differential techniques used in this work. Second, we seek to develop new probes of the photoionization process, e.g., fluorescence polarization spectroscopy, which will yield new types of information. Third, this project produces data crucial for testing theoretical predictions and, thus, contributes to the development of realistic theories of atomic and molecular photoionization. Fourth, the data produced by this project contributes to characterizing all the pathways by which radiation interacts with matter, and hence contributes to the macroscopic modeling of such interactions.

4. Scientific and Technical Approach

This program utilized three experimental approaches: First, the main effort involves measuring triply differential photoelectron cross sections using synchrotron radiation. Thus, the intensity of photoelectrons ejected from atoms and molecules are measured as a function of three

independent parameters — the wavelength of the incident synchrotron radiation, the kinetic energy of the photoelectron, and the ejection angle relative to the polarization direction of the light. Second, the polarization of fluorescence from excited ionic states produced by photoionization is measured as a function of the wavelength of the incident light. Third, photoelectrons and photoions are measured in coincidence to correlate a photoelectron energy with a component species in a mixture (used in the last period to record the first photoelectron spectra of an atomic cluster larger than dimer) or, alternatively, to correlate an ionic fragment with a particular state of the parent photoion.

##### 5. Progress

The scientific accomplishments of this program during the last contract period are reflected, in part, in the papers, abstracts of contributed talks, and invited lectures listed in Section 6. In particular, papers 14 and 15 were previously submitted, but have appeared in this contract period. Papers 16-26 are new in this contract period. In addition, abstracts 22-30 and invited talks 20-25 were added during this contract period.

The highlights of the past year, not all of which are represented by the papers, abstracts, and talks, can be summarized in six categories: First, and most significant for the future of the program, we have devoted the major effort in this period to the completion of a new generation double electron spectrometer system for triply differential photoionization studies. This new system has been under development for nearly two years and will yield two to three orders of magnitude improvement in sensitivity/resolution over our previous instrument and others that have appeared since our initial work in this field. This is crucial for addressing the most significant problems in the field now and in the future. The system has been completed and tested with channeltron detectors, demonstrating a resolution of <0.010 eV, which is approximately ten times higher resolution than is currently available from instruments being used with synchrotron radiation. Installation of existing area detectors will take place after a data taking period and will vastly improve the sensitivity of our studies. The fully optimized system will allow exploration of phenomena which is beyond the reach of existing instruments. Second, several papers have been prepared describing triply differential photoelectron studies of molecular photoionization dynamics, including shape resonances (papers 15, 17, 24, 26), autoionizing resonances (papers 14, 17, 24, 26), and strong continuum-continuum coupling (paper 15). Third, fluorescence polarization studies, pioneered by this project (paper 3), have continued with a major experimental run resulting in several new studies. These include spectroscopic uses of fluorescence polarization (paper 19), probes of electric field effects on photoionization, etc. Most of this work is still being prepared for publication. Fourth, a new constant-photoelectron-energy mode was utilized for studying photoionization processes in the near threshold region in C<sub>2</sub>H<sub>2</sub> (paper 16). Fifth, paper 21 is a major review article describing progress in the study of photoionization dynamics of small molecules. Sixth, other studies (papers 18, 22, 25) have provided new molecular photoionization data on various topics.

6. Publications

The following pages list papers, abstracts of contributed talks, and invited talks resulting from this program. Papers 14 and 15 were submitted earlier, but were published during this contract period. Papers 16-26, abstracts 22-30, and invited talks 20-25 are new in this period.

PAPERS

1. B. E. Cole, D. L. Ederer, R. Stockbauer, K. Codling, A. C. Parr, J. B. West, E. D. Poliakoff, and J. L. Dehmer, "Wavelength and Vibrational-State Dependence of Photoelectron Angular Distributions. Resonance Effects in  $5\sigma$  Photoionization of CO," *J. Chem. Phys.* 72, 6308 (1980).
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10. D. M. P. Holland, A. C. Parr, D. L. Ederer, J. L. Dehmer, and J. B. West, "The Angular Distribution Parameters of Argon, Krypton, and Xenon for Use in Calibration of Electron Spectrometers," *Nucl. Instr. and Meth.* 195, 331 (1982).
11. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. Stockbauer, "Photoelectron-Photoion Coincidence Spectroscopy of Gas-Phase Clusters," *J. Chem. Phys.* 76, 5214 (1982).

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12. A. C. Parr, D. L. Ederer, J. B. West, D. M. P. Holland, and J. L. Dehmer, "Triply Differential Photoelectron Studies of Non-Franck-Condon Behavior in the Photoionization of Acetylene," *J. Chem. Phys.* 76, 4349 (1982).
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15. J. L. Dehmer, A. C. Parr, S. Wallace, and D. Dill, "Photoelectron Branching Ratios and Angular Distributions for the Valence Levels of  $\text{SF}_6$  in the Range  $16\text{eV} < h\nu < 30\text{eV}$ ," *Phys. Rev. A* 26, 3283 (1982).
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20. A. C. Parr, S. H. Southworth, J. L. Dehmer, and D. M. P. Holland, "Photoelectron Spectrometer for High Resolution Angular Resolved Studies," *Nucl. Instr. and Meth.*, in press.
21. J. L. Dehmer, D. Dill, and A. C. Parr, "Photoionization Dynamics of Small Molecules" in Photophysics and Photochemistry in the Vacuum Ultraviolet, edited by S. McGlynn, G. Findley, and R. Huebner (D. Reidel Publ., Dordrecht, Holland, 1983), in press.
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PAPERS, Continued

24. D. M. P. Holland, A. C. Parr, D. L. Ederer, J. B. West, and J. L. Dehmer, "Triply Differential Photoelectron Studies of the Four Outermost Valence Orbitals of Cyanogen," Int. J. Mass Spect. Ion Phys., in press.
25. D. M. P. Holland, A. C. Parr, and J. L. Dehmer, "Photoelectron Asymmetry Parameters and Branching Ratios for Sulphur Dioxide in the Photon Energy Range 14 to 25 eV," J. Electron Spectrosc., to be submitted.
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2. K. Codling, J. B. West, A. C. Parr, J. L. Dehmer, B. E. Cole, D. L. Ederer, and R. L. Stockbauer, "Partial Cross Sections, Vibrational Branching Ratios, and Angular Distributions in the 570-600 Å Window Resonance in O<sub>2</sub>," *ibid.* p. II-14.
3. R. Stockbauer, A. C. Parr, J. L. Dehmer, B. E. Cole, D. L. Ederer, J. B. West, and K. Codling, "Perturbation of Vibrational Intensities and Angular Distributions by Autoionization in Molecular Photoionization," *ibid.*, p. II-15.
4. E. D. Poliakoff, J. L. Dehmer, A. C. Parr, D. Dill, K. H. Jackson, and R. N. Zare, "Polarized Fluorescence Excitation Spectroscopy of N<sub>2</sub>," *ibid.*, p. II-25.
5. J. L. Dehmer, A. C. Parr, J. B. West, K. Codling, D. L. Ederer, B. E. Cole, E. D. Poliakoff, and R. Stockbauer, "Effects of Shape Resonances on Vibrational Branching Ratios and Photoelectron Angular Distributions in Molecular Photoionization," *ibid.*, p. II-86.
6. A. C. Parr, R. L. Stockbauer, K. Codling, J. B. West, and J. L. Dehmer, "Photoelectron Branching Ratios and Angular Distributions in the Region of the 3s3p<sup>6</sup>4p <sup>1</sup>P<sub>1</sub><sup>o</sup> Resonance in Ar and the 5s5p<sup>6</sup>6p <sup>1</sup>P<sub>1</sub><sup>o</sup> Resonance in Xe," Annual DEAP Meeting, 1-3 December 1980, Los Angeles, CA, Bull. Am. Phys. Soc. 25, 1131 (1980).
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8. E. D. Poliakoff, J. L. Dehmer, D. Dill, A. C. Parr, K. H. Jackson, and R. N. Zare, "Polarization of Fluorescence Following Molecular Photoionization," *ibid.*, p. 1136.
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10. R. Stockbauer, A. C. Parr, B. E. Cole, D. L. Ederer, J. Dehmer, J. West, and K. Codling, "Effects of Two-Electron Resonances in Photoelectron Energy and Angular Distributions," presented orally at the Gordon Research Conference on Electron Spectroscopy, Wolfeboro, NH, July 1980 (no abstract available).

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17. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. Stockbauer, "Photoelectron-Photoion Coincidence Spectroscopy of Gas-Phase Clusters," *ibid.*, p. 1322.
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19. A. C. Parr and H. M. Rosenstock, "Resonance, Autoionization, and Kinetic Effects in Photoionization," (invited talk) Annual Meeting of the American Society for Mass Spectrometry, 7-11 June 1982, Honolulu, Hawaii, Book of Abstracts.
20. J. L. Dehmer, D. Dill, and A. C. Parr, "Molecular Photoionization Dynamics with Emphasis on Shape and Autoionizing Resonances," Abstract of Invited Lecture at the NATO Advanced Study Institute on Photophysics and Photochemistry in the Vacuum Ultraviolet, 15-28 August 1982, Lake Geneva, Wisconsin, Book of Abstracts.

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21. A. C. Parr and J. L. Dehmer, "Photoelectron Spectrometer for High Resolution Angular Resolved Studies," International Conference on X-Ray and VUV Synchrotron Radiation Instrumentation, 9-13 August 1982, Hamburg, W. Germany, Book of Abstracts.
22. D. L. Ederer, R. P. Madden, A. C. Parr, G. Rakowsky, E. B. Saloman, J. Cooper, R. Stockbauer, T. E. Madey, and J. L. Dehmer, "An Overview of Research at NBS Using Synchrotron Radiation at SURF-II," Seventh Conf. on the Application of Accelerators in Research and Industry, 8-10 November 1982, Denton, TX, Bull. Am. Phys. Soc. 27, 809 (1982).
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24. J. L. Dehmer, A. C. Parr, S. H. Southworth, and D. M. P. Holland, "Triply Differential Photoelectron Studies of Autoionization and Shape-Resonance Effects in Molecular Photoionization," Annual DEAP Meeting, 23-25 May 1983, Boulder, CO, Bull. Am. Phys. Soc. 28, 809 (1983).
25. E. D. Poliakoff, J. L. Dehmer, A. C. Parr, and G. E. Leroi, "Fluorescence Polarization as a Probe of Molecular Autoionization," *ibid.*, p. 809.
26. J. L. Dehmer, A. C. Parr, S. H. Southworth, and D. M. P. Holland, "Triply Differential Photoelectron Studies of Resonant Molecular Photoionization," XIII International Conference on the Physics of Electronic and Atomic Collisions, Berlin, W. Germany, 27 July - 2 August 1983, Book of Abstracts.
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28. E. D. Poliakoff, J. L. Dehmer, A. C. Parr, and G. E. Leroi, "Fluorescence Excitation Studies of Molecular Photoionization in the Presence of External Electric Fields," VII International Conference on Vacuum Ultraviolet Radiation Physics, Jerusalem, Israel, 8-12 August 1982, Book of Abstracts; also in Ann. Israel Phys. Soc. 6, XX (1983).
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30. A. C. Parr and R. Stockbauer, "Photoelectron Spectroscopy Studies in Recent Years at NBS," Annual Meeting of the American Society for Mass Spectrometry, Boston, MA, 9-13 May 1983, Book of Abstracts.

INVITED TALKS, COLLOQUIA, AND SEMINARS

1. J. L. Dehmer and Dan Dill, "Shape Resonances in Molecular Photoionization," Plenary talk presented at Molecular Spectroscopy and Dynamics with Synchrotron Radiation-A European Workshop, Maria Laach, West Germany, September 29-October 1, 1980.
2. A. C. Parr, "Current Research at NBS Using Synchrotron Radiation at SURF-II," Invited talk presented at the Sixth Conference on the Application of Accelerators in Research and Industry, Denton, TX, November 3-5, 1980.
3. J. L. Dehmer, "Potpourri of Current and Future Studies of Molecular Photoionization-Synchrotron Radiation, Supersonic Jets, and Multiphoton Ionization," Chemistry Department Colloquium, Boston University, Boston, MA, 13 April 1981.
4. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Photoelectron-Photoion Coincidence Spectroscopy of Atomic Clusters and Fluorescence Polarization Analysis," Atomic and Molecular Science Seminar, Argonne National Laboratory, Argonne, Illinois, 6 May 1981.
5. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Molecular Spectroscopy Division Seminar, National Bureau of Standards, Gaithersburg, MD, 21 May 1981.
6. E. D. Poliakoff, "Alignment of Molecular Ions Produced by Photoionization," Seminar on Collision Experiments in Their Theoretical Frame (Fano Workshop), The University of Chicago, Chicago, IL, 23 May 1981.
7. A. C. Parr, "Status of Programs at NBS SURF-II," National Synchrotron Instrumentation Conference, Cornell University, 15-17 July 1981.
8. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Physics Department Colloquium, Georgetown University, Washington, D.C. 6 November 1981.
9. A. C. Parr, "Triply Differential Photoelectron Spectrometry of Atoms and Simple Molecules," Center Colloquium, Center for Absolute Physical Quantities, National Bureau of Standards, January 6, 1982.
10. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Brookhaven National Laboratory, December 2, 1981.
11. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Exxon Research Laboratory, January, 1982.
12. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Department of Chemistry, Dartmouth University, January, 1982.

INVITED TALKS, COLLOQUIA, AND SEMINARS, Continued

13. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Department of Chemistry, Boston University, January 1982.
14. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Department of Chemistry, University of Pennsylvania, February, 1982.
15. A. C. Parr and H. M. Rosenstock, "Resonance, Autoionization, and Kinetic Effects in Photoionization," Invited talk presented at the 30th Annual Conf. on Mass Spectrometry and Allied Topics, Honolulu, June 6-11, 1982.
16. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Department of Physics Colloquium, University of Alabama, March 31, 1982.
17. J. L. Dehmer, "Molecular Photoionization Dynamics - Progress and Prospects," Physics Colloquium, University of Chicago, May 27, 1982.
18. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," California Institute of Technology, June 14, 1982.
19. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," University of California, Santa Barbara, June 15, 1982.
20. J. L. Dehmer, "Overview of Experimental and Theoretical Studies of Resonance Processes in Molecular Photoionization by Single-Photon and Multiphoton Excitation," Gordon Research Conference on Electron Spectroscopy, Wolfeboro, New Hampshire, 19 July, 1982.
21. J. L. Dehmer, D. Dill, and A. C. Parr, "Photoionization Dynamics of Small Molecules," NATO Advanced Study Institute on Photophysics and Photochemistry in the Vacuum Ultraviolet, Lake Geneva, WI, 15-28 August 1982.
22. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Uppsala University, 17 August 1982.
23. J. L. Dehmer, "Resonant Processes in Molecular Photoionization," Meeting of the American Physical Society (Division of Condensed Matter Physics Symposium), Los Angeles, CA, 24 March 1983.
24. A. C. Parr, "Studies of Ion Fragmentation and Molecular Photoionization Using Photoelectron Spectroscopy," JILA Colloquium, Boulder, CO, 8 April 1983.
25. A. C. Parr and R. Stockbauer, "Photoelectron Spectroscopy Studies in Recent Years at NBS," Annual Meeting of the American Society for Mass Spectrometry, Boston, MA, 12 May 1983.

7. Extenuating Circumstances

None.

8. Unspent Funds

None will remain unspent at the end of the current contract period.

9. Graduate Students Receiving Degrees

None.

10. Other Federal Contract Support

During this period, J. L. Dehmer was a co-principal investigator for Office of Naval Research Contract N00014-83-F-0001, "Selectivity of Multiphoton Processes," 10/1/82 - 9/30/83.

During this period, R. Stockbauer was a co-principal investigator for Office of Naval Research Contract N00014-83-F-0002, "Characterization of Surface Bonding Using Photon and Electron Stimulated Desorption," 10/1/82 - 9/30/83.

